

The Thermal Conductivity of R115 in the Critical Region¹

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By means of the transient hot-wire method, the thermal conductivity of refrigerant R115 was measured over a wide region around the thermodynamic critical point. The enhancement of thermal conductivity was clearly observed at states close to the critical point. The thermal conductivity obtained for saturated liquid was compared to data by other authors with satisfactory agreement. For the thermal conductivity including the critical enhancement, equations are given according to the suggestion by J. V. Sengers. Comparison with measured data shows systematic deviations, with theoretical values being larger except for temperatures very close to the critical and densities above the critical.

KEY WORDS: critical point; enhancement; fluorochlorohydrocarbons; hot-wire method; refrigerant R115; thermal conductivity; transient method.

1. INTRODUCTION

Properties of refrigerants are in general well investigated at low temperatures, as these give the interesting range for refrigeration. At high temperatures, e.g., near the thermodynamic critical point, experimental investigations are rather scarce and the few results often deviate from each other. Here the thermal conductivity of R115 (C₂ClF₅), supplied by Hoechst AG/FRG, is measured within a wide range of temperatures, pressures, and densities, listed in Table I. Its purity is 99.994%.

¹ Paper presented at the Tenth Symposium on Thermophysical Properties, June 20–23, 1988, Gaithersburg, Maryland, U.S.A.

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Table I. Experimental Range and Critical Data [1]

	Experimental range	Critical data
Temperature (K)	$290 < T < 370$	$T_c = 353.15$
Pressure (bar)	$2 < p < 70$	$p_c = 31.26$
Density ($\text{kg} \cdot \text{m}^{-3}$)	$13 < \rho < 1400$	$\rho_c = 591$

2. EXPERIMENTS

2.1. Apparatus

The measurements were carried out applying the transient hot-wire method [2–6] in an apparatus described in Ref. 7. The characteristics of this apparatus are compiled in Table II.

2.2. Procedure

As described in Ref. 9 the governing equation is

$$\Delta T_{id} = \Delta T_w + \sum_i \delta T_i = \frac{q_1}{4\pi\lambda} \ln \left[\frac{4a\tau}{r^2 C} \right] \quad (1)$$

with $\Delta T_w = T(\tau) - T_0$ being the temperature rise of the electrically heated wire from the initial temperature T_0 during the time τ , q_1 being the heat flux per unit length of the wire, λ and a the thermal conductivity and diffusivity of the fluid, r the radius of the wire, C a constant, and δT_i the

Table II. Characteristics of the Hot-Wire Apparatus Containing One Measurement and One Compensation Cell [7]

Cell	
Height	230 mm
Radius	2.5 mm for liquids, 8 mm for gases
Wire	
Material	Platinum
Radius	0.0085 mm
Length	154.1 ± 0.1 mm for measurement 63.8 ± 0.1 mm for compensation
Resistance (at 20.6°C)	$92.26 \pm 0.05 \Omega$ for measurement $38.23 \pm 0.02 \Omega$ for compensation

Table III. Thermal Conductivity of R115 at Subcritical Temperatures^a

$T_{nom}(K)$		p (bar)	ρ (kg · m ⁻³)	λ (W · m ⁻¹ · K ⁻¹)
289.65	*	7.08	1324	0.05314
290.15	*	7.27	1321	0.05311
294.15		60.76	1361	0.05635
		39.96	1340	0.05481
		31.26	1332	0.05431
		20.56	1321	0.05365
		12.36	1314	0.05331
		* 7.90	1305	0.05212
			5.45	39.2
		1.96	12.7	0.0131
301.75	*	9.80	1265	0.04998
304.15		59.96	1314	0.05444
		47.97	1302	0.05264
		31.26	1283	0.05168
		20.56	1270	0.05078
		* 10.28	1255	0.04953
		* 10.27	82.6	0.01496
305.65	*	10.81	1244	0.04864
309.65	*	11.94	1222	0.04794
310.75	*	12.23	1216	0.04682
314.15		50.06	1257	0.05093
		40.0	1244	0.04936
		31.26	1230	0.04835
		20.56	1212	0.04729
		* 13.15	1200	0.0464
		13.15	106.6	0.0155
		9.44	67.8	0.01495
		5.76	37.9	0.01421
		2.07	12.9	0.01365
	323.65		64.14	1230
		51.76	1211	0.04728
		31.26	1173	0.04534
		24.17	1158	0.04477
		20.56	1147	0.0437
		* 16.58	1135	0.0426
		16.08	133.3	0.01648
		9.88	67.7	0.01541
		5.96	37.9	0.01461
		2.17	12.9	0.01389

^a An asterisk indicates the saturation state.

Table III. (Continued)

$T_{\text{nom}}(K)$	p (bar)	ρ ($\text{kg} \cdot \text{m}^{-3}$)	λ ($\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$)	
333.65	54.97	1162	0.04615	
	49.97	1151	0.04528	
	36.96	1124	0.0435	
	31.26	1103	0.0423	
	23.95	1077	0.04065	
	21.45	1067	0.04055	
	*	20.56	1058	0.03953
		16.96	131.1	0.01669
		10.27	67.5	0.01559
		6.17	37.9	0.01479
		2.26	12.9	0.01439
	343.65	62.17	1119	0.04365
		52.96	1096	0.0426
43.67		1068	0.04143	
40.76		1058	0.04025	
33.45		1026	0.0393	
31.26		1018	0.0382	
28.97		1006	0.03774	
26.56		992	0.0366	
*		25.5	266	0.02062
		22.3	190	0.0185
		17.77	130.5	0.01753
		10.76	67.9	0.016
		6.37	37.6	0.01502
		2.36	12.9	0.0146

various additive corrections. The correction terms δT_i in Eq. (1) have been detailed in Refs. 8 and 9. A first correction, δT_1 , accounts for the finite heat capacity of the wire and has been given by Healy [9]. In the present measurements this correction has been applied. A correction δT_2 arises from the effects of axial heat conduction near the supports. These end effects have been compensated in the present experiments by using two wires [8]. Further corrections, which arise, e.g., from the presence of an outer boundary to the fluid or from radiation, have been neglected in the present work due to their small amounts.

The thermal conductivity can be calculated from

$$\lambda = \frac{q_1}{4\pi l} \frac{d(\Delta T_{id})}{d(\ln \tau)} \quad (2)$$

where $d(\Delta T_{id})/d(\ln \tau)$ is obtained from consecutively recorded temperatures after correcting if necessary. This temperature increase is linear as long as

only heat conduction determines the heat dissipation from the wire during a certain time interval. Measurements were performed at subcritical and supercritical isothermal conditions (Tables III and IV). Also, measurements were taken at the saturation state.

The temperature of the hot-wire bath was measured by a Hg thermometer with an accuracy of ± 0.1 K; the transient temperature of the wire is transferred into a voltage change of a precision Wheatstone bridge and recorded by a computerized data acquisition system. The pressure was measured by a calibrated pressure gauge with an accuracy of ± 0.05 bar.

Densities were calculated for $\rho < 400 \text{ kg} \cdot \text{m}^{-3}$ from the equation of state proposed in Ref. 10 and for $\rho > 900 \text{ kg} \cdot \text{m}^{-3}$ from that proposed in Ref. 11. In the critical region, the densities could also be directly determined with our apparatus and a calibrated cylinder/spindle arrangement. If the filling of the cylinder is displaced, the density in the pressure cell increases by $\Delta\rho = 52.5 \pm 0.5 \text{ kg} \cdot \text{m}^{-3}$. The accuracy achieved was estimated to be 3% for the critical region and 2% for the other regions.

3. RESULTS

The results are shown in Fig. 1. For liquid R115, in the subcooled region ($p > p_{\text{saturation}}$ for $T = \text{const}$), the pressure effect upon the thermal conductivity is small. It is more pronounced for saturation conditions; the

Table IV. Thermal Conductivity of R115 at Supercritical Temperatures

T (K)	p (bar)	ρ ($\text{kg} \cdot \text{m}^{-3}$)	λ ($\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$)
353.65	2.4	12.9	0.0150
	2.5	13.5	0.0155
	11.04	66.9	0.0166
	19.12	134	0.0176
	23.65	187	0.01881
	26.81	239	0.0198
	27.30	252	0.0209
	28.93	292	0.0220
	30.30	344	0.0262
	30.96	397	0.0367
	31.07	450	0.03813
	31.11	502	0.0499
	31.17	528	0.0522
	32.7	766	0.0398
	38.97	819	0.0383
	41.36	977	0.0378
	49.97	1027	0.0401
68.97	1080	0.0414	

Table IV. (Continued)

T (K)	p (bar)	ρ (kg · m ⁻³)	λ (W · m ⁻¹ · K ⁻¹)
355.40	27.63	248	0.0209
	28.82	275	0.0224
	29.68	300	0.0250
	31.05	354	0.0275
	31.79	406	0.0301
	32.22	460	0.0367
	32.35	486	0.0386
	32.44	513	0.0408
	32.55	539	0.0428
	32.63	565	0.04675
	32.64	592	0.0548
	32.68	607	0.0557
	32.76	644	0.0545
	32.91	671	0.0469
	32.95	698	0.0408
	33.37	751	0.0394
	33.67	777	0.0367
	34.66	830	0.0365
	37.07	883	0.03711
357.65	24.08	185	0.0193
	26.01	212	0.0206
	27.54	238	0.0224
	29.97	291	0.0242
	31.51	344	0.0253
	32.55	397	0.0314
	33.15	450	0.03525
	33.61	502	0.0386
	33.69	528	0.0395
	33.83	555	0.04216
	33.94	581	0.0431
	34.12	607	0.0441
	34.22	633	0.0426
	357.65	34.35	660
34.5		686	0.04059
34.63		713	0.03975
34.77		739	0.0383
35.04		766	0.0375
36.35		819	0.03623
37.07		845	0.0366
38.46		872	0.0374
44.86		952	0.03816

Table IV. (Continued)

T (K)	p (bar)	ρ ($\text{kg} \cdot \text{m}^{-3}$)	λ ($\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$)
361.65	20.83	142	0.01895
	25.36	195	0.0193
	27.16	222	0.02155
	28.92	250	0.0233
	32.31	327	0.0256
	34.25	405	0.0291
	34.72	433	0.0321
	35.11	460	0.03267
	35.42	486	0.03426
	35.77	513	0.03538
	36.05	539	0.0358
	36.28	565	0.03645
	36.51	592	0.03698
	36.68	607	0.03814
	36.79	618	0.03764
	37.25	671	0.03753
	37.49	698	0.0374
38.59	777	0.0359	
40.03	830	0.0368	
43.01	883	0.0367	
49.96	961	0.0375	
363.65	35.67	448	0.0300
	36.3	475	0.0327
	36.67	508	0.03435
	37.32	555	0.03504
	37.65	581	0.03629
	37.93	607	0.03689
	38.25	633	0.0365
	38.56	660	0.0365
	39.15	713	0.0361
	39.94	766	0.0358
	43.96	872	0.0365
49.16	924	0.0369	
68.87	1030	0.0407	
368.65	38.72	476	0.0311
	39.78	529	0.0330
	40.67	581	0.0340
	41.17	607	0.0343
	43.45	738	0.0350
	45.94	819	0.0362

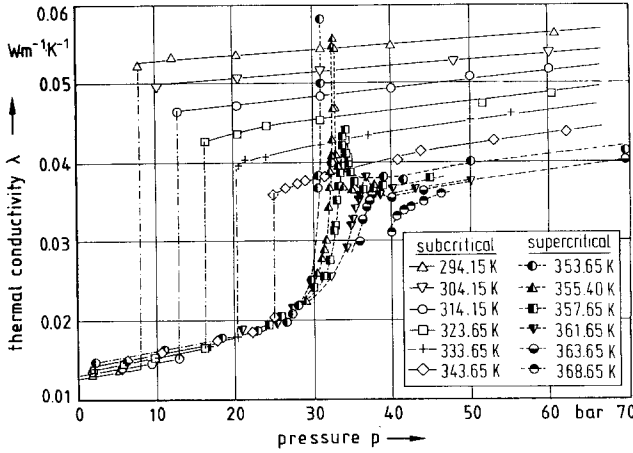


Fig. 1. Thermal conductivity measured for R115 in the liquid and gaseous state.

thermal conductivity drops to quite small values (about 1/5) for gaseous R115. When the critical region is approached the differences in λ between liquid and gaseous state diminish and the enhancement in thermal conductivity near the critical point is observed: sharp peaks appear.

The measured data are also presented in Tables III and IV. In order to correlate the measured conductivities to the same isotherms, T_{nom} is introduced and small temperature corrections had to be made according to

$$\lambda(T_{nom}, \rho) = \lambda(T, \rho) + (\partial\lambda/\partial T)_\rho (T_{nom} - T) \tag{3}$$

from Eq. (10) with $(\partial\lambda/\partial T)_\rho = 3.187 \cdot 10^{-5} \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ from Eq. (9).

4. ACCURACY

The accuracy of measurements with our hot-wire apparatus was tested with toluene at atmospheric pressure [7] and proved to be within $\pm 1.6\%$. For R115 measurements at higher pressures and temperatures but far away from the critical point, the accuracy was evaluated as $\pm 2.33\%$ for 95% of the data. For measurements in the critical region, some problems appear.

- (a) The onset of natural convection in the fluid occurs after a very short heating time.
- (b) As a consequence the effect of the different heat capacities of the wire and the fluid is not negligible.
- (c) The fluid properties change strongly with temperature.

The experimental results show that the onset of natural convection occurs about 0.2 s after heating starts when $T - T_c = 2.25$ K at the critical isochore. The length of the time interval increases when this temperature difference increases. Due to the short time, the determination of the slope $d(\Delta T_{id})/d(\ln \tau)$ becomes inaccurate. The slope is usually obtained from about 40 to 60 pairs of data which are processed by rms analysis. Near the critical point, only a few pairs are available. The accuracy, which is about $\pm 0.2\%$ for data far away from the critical region, drops to 4% when $T - T_c = 2.25$ K at the critical isochore. Data points within $|T - T_c| < 2$ K and $|\rho - \rho_c| < 200 \text{ kg} \cdot \text{m}^{-3}$ were measured (Fig. 1) but their accuracy is very poor and they are not taken for further treatment (Fig. 3). The effect of different heat capacities can be corrected [9]; the results show that another drop in accuracy occurs with 1 to 4% for the short heating times. The effect of temperature-dependent properties—which actually restricts the applicability of Eq. (1) principally—was approximated by solving Eq. (4) numerically with variable properties:

$$\rho c_p \frac{\partial(\Delta T)}{\partial \tau} = \frac{1}{r} \frac{\partial}{\partial r} \left[\lambda r \frac{\partial(\Delta T)}{\partial r} \right] \quad (4)$$

It can be shown by a power-law consideration near critical conditions that, for a small temperature rise and $|T - T_c| > 2$ K, a linear temperature dependence may be assumed

$$\rho c_p = (1 + \beta^* \Delta T)(\rho c_p)_0 \quad (5a)$$

$$\lambda = (1 + \gamma^* \Delta T)\lambda_0 \quad (5b)$$

with $|\beta^*| \leq 0.6$ and $|\gamma^*| \leq 0.32$, the index 0 indicating the starting state. In the numerical solution values of $|\beta^*|$ and $|\gamma^*|$ were tested up to 0.8. Maximum errors were found to be within $\pm 3\%$ for the reference temperature: $T = T_0 + 0.5 [\Delta T(\tau_1) + \Delta T(\tau_2)]$, where τ_1 and τ_2 are the beginning and end times of the measurement. The density stratification in this arrangement has been studied and the maximum relative deviation of the density along the wire was found to be $|\Delta \rho / \rho_c| < 0.5\%$ for $|T - T_c| \geq 2$ K. That means the deviation of the density is within the accuracy of the density measurement. Thus the overall accuracy for our measurements of λ near the critical point, but with $|T - T_c| > 2$ K, is within 5 to 11%.

5. COMPARISON

Only a few measurements for the thermal conductivity of R115 at saturation state are reported in literature. A comparison between our

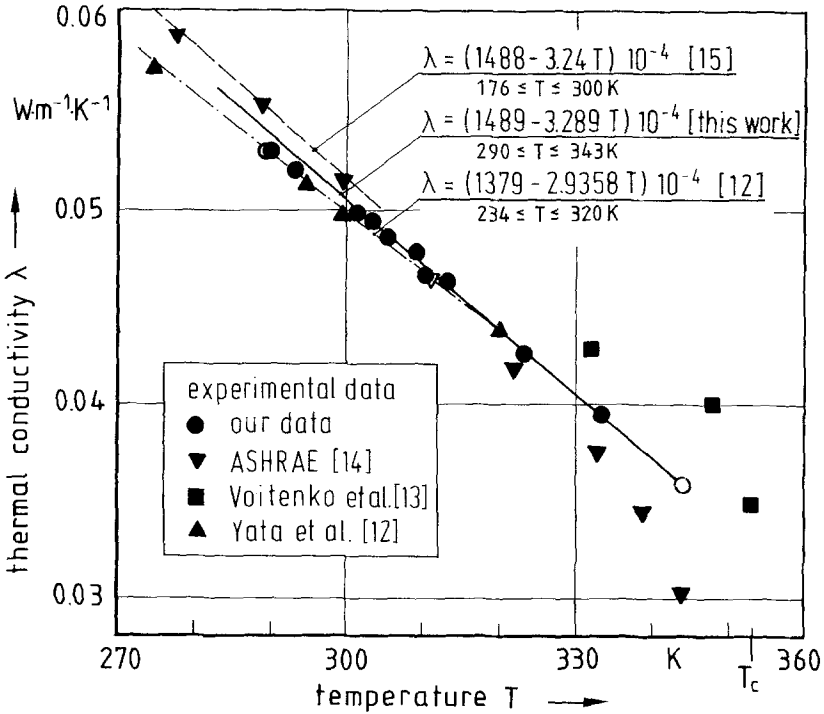


Fig. 2. Thermal conductivity measured for saturated liquid by various authors.

results and those from the literature is presented in Fig. 2. There is good agreement with results by Yata et al. [12]. The results given in Ref. 13 are 25% higher than ours and those given in Ref. 14 are 15% lower. The thermal conductivity λ , $W \cdot m^{-1} \cdot K^{-1}$, for saturated liquid as calculated from the equation

$$\lambda = (1489 - 3.289 T) 10^{-4} \tag{6}$$

for $290 < T < 343$ K, differs from that of Ref. 15 by 2.4 to 4.3% (ours is lower) and that of Ref. 12 by 1.5% (ours is higher).

6. EQUATIONS

Following a suggestion by Sengers [16] the thermal conductivity of fluids near their critical point can be given as the sum of three terms,

$$\lambda(\rho, T) = \lambda(0, T) + \lambda(\rho) + \Delta\lambda_c(\rho, T) \tag{7}$$

with $\lambda(0, T)$ being the thermal conductivity of the dilute gas (with a very low density), $\lambda(\rho)$ the excess thermal conductivity (which depends on the density), and $\Delta\lambda_c(\rho, T)$ the so-called critical enhancement. The sum

$$\lambda(0, T) + \lambda(\rho) = \lambda_B(\rho, T) \quad (8)$$

is called the “background thermal conductivity.” This is a weak function of temperature and a somewhat stronger function of density.

6.1. Background Thermal Conductivity

This is the thermal conductivity of the fluid far away from the critical point, where the critical enhancement is not observed. The thermal conductivity at very low density $\lambda(0, T)$ is obtained from our measurements as

$$\lambda(0, T) = (34.75 + 0.3187 T) 10^{-4} \quad (9)$$

with T in K, and $\lambda(0, T)$ in $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ for the temperature range $293 < T < 363$ K. This equation was obtained from different temperature data extrapolated to $\rho = 0$.

The background thermal conductivity for R115 was obtained as the polynomial expression

$$\lambda_B(\rho, T) = \lambda(0, T) + a_1\rho + a_2\rho^2 + a_3\rho^3 \quad (10)$$

with $\lambda(0, T)$ from Eq. (9) and

$$a_1 = 24.363 \times 10^{-6}$$

$$a_2 = -17.695 \times 10^{-9}$$

$$a_3 = 17.273 \times 10^{-12}$$

for T in K, ρ in $\text{kg} \cdot \text{m}^{-3}$, and λ in $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$. For this equation all thermal conductivities measured at states far away from the critical region, at $\rho < 270 \text{ kg} \cdot \text{m}^{-3}$ or $\rho > 960 \text{ kg} \cdot \text{m}^{-3}$, were used in a rms analysis. The standard deviation between our experimental data (away from the critical region) and the background thermal conductivity as calculated from Eqs. (9) and (10) is $\pm 1.19\%$.

The curve for λ_B is shown together with measured data in Fig. 3.

6.2. Critical Enhancement

The theory of dynamic critical phenomena predicts that the critical enhancement $\Delta\lambda_c$ will strongly increase in approach to the critical point (for $\rho \rightarrow \rho_c$, $T \rightarrow T_c$).

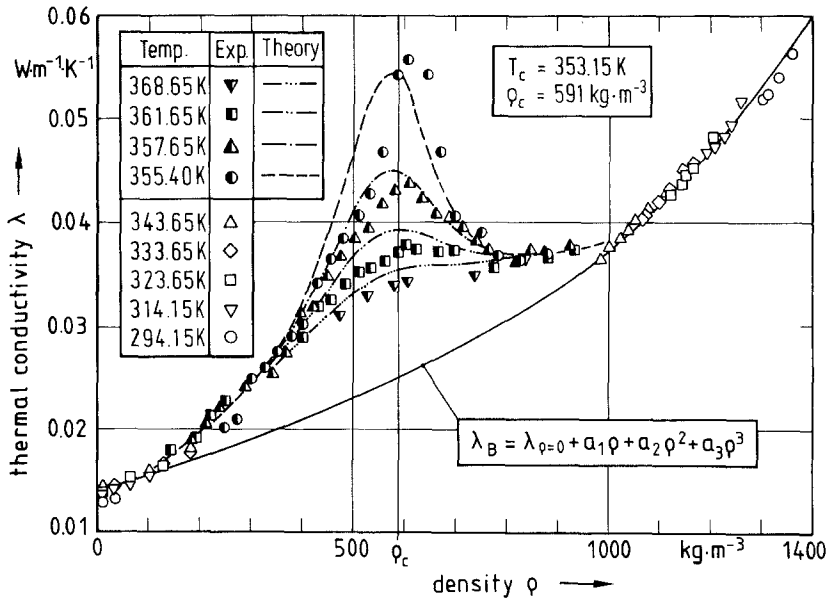


Fig. 3. Thermal conductivity as measured and calculated.

Figure 3 shows this effect on the thermal conductivity measured for densities $140 < \rho < 950 \text{ kg} \cdot \text{m}^{-3}$ and supercritical temperatures. Close to the critical density a distinct enhancement $\Delta\lambda_c$ is superposed to the curve for the background thermal conductivity λ_B .

Basu and Sengers [17] proposed an equation for the critical enhancement, which is built up from three factors:

$$\Delta\lambda_c = A \Delta\lambda'_c F(\Delta T^*, \Delta\rho^*) \quad (11)$$

The *first* factor, A , is a constant. Several values ranging between 1.15 and 1.22 have been reported in Ref. 17. Here $A = 1.2$ was chosen for a better fit of experimental data.

The *second* factor, $\Delta\lambda'_c$, can be written as

$$\Delta\lambda'_c(\rho, T) = \frac{k_B p_c}{6\pi\eta\xi} \left[\frac{\partial p^*}{\partial T^*} \right]_{\rho}^2 \left[\frac{T^*}{\rho^*} \right]^2 \chi_T^* \quad (12)$$

where $k_B = 1.38 \cdot 10^{-23} \text{ J} \cdot \text{K}^{-1}$ is Boltzmann's constant, η is the shear viscosity, ξ and χ_T^* are the correlation length and symmetrized isothermal compressibility, respectively, and $\rho^* = \rho/\rho_c$, $T^* = T/T_c$, and $p^* = p/p_c$ are reduced properties. The equation for $\Delta\lambda'_c$ refers to the asymptotic behavior

of $\Delta\lambda_c$ near the critical point. Far away from the critical point, the critical enhancement contributions should vanish.

The *third* factor in Eq. (11), $F(\Delta T^*, \Delta\rho^*)$, is a damping function [18] and has the form

$$F(\Delta T^*, \Delta\rho^*) = \left[\frac{\rho}{\rho_c} \right]^n \exp[-A(\Delta T^*)^2 - B(\Delta\rho^*)^4] \quad (13)$$

with $\Delta T^* = (T - T_c)/T_c$ and $\Delta\rho^* = (\rho - \rho_c)/\rho_c$. The coefficients A and B and the exponent n are treated as adjustable constants, being chosen as $A = 18.66$, $B = 1.0$, and $n = 0.5$, just as used by Basu and Sengers [17] for water. For an accurate representation of the equilibrium properties in the critical region—used in Eq. (12)—we apply relations which are compiled by Basu and Sengers [19].

In Fig. 3 the thermal conductivity of R115 calculated from Eqs. (7) to (13) is compared with the experimental data in the critical region at four supercritical temperatures. The agreement between theoretical and experimental data proves to be satisfactory. Peaks of the thermal conductivity, measured at constant temperature, are slightly shifted to densities above the critical. A similar result has been reported previously, e.g., by Sengers et al. [20] (for H_2O) and by Prasad and Venart [21] (for ethane).

It is shown in Fig. 3 that above 357.65 K the experimental data are systematically smaller than the theoretical data. This may be due to the determination error of the background thermal conductivity. Because λ_B is obtained by interpolation from the experimental data taken far away from the critical density, the values of λ_B at the critical density might be slightly too large.

7. CONCLUSIONS

The thermal conductivity of R115 has been investigated experimentally as well as theoretically in a wide region around the critical point.

The *experiments*—carried out by means of the transient hot-wire technique—show the following.

- (a) The method may be applied with an accuracy of about $\pm 2.3\%$ at states far away from the critical point and of about $\pm 10\%$ close to it (with $|T - T_c| > 2$ K).
- (b) The thermal conductivity of saturated liquid R115 is in good agreement with measurements reported in the literature at temperatures far below the critical, $(T_c - T) > 25$ K; at temperatures $(T_c - T) < 25$ K, deviations increase.

- (c) A distinct so-called "critical enhancement" of the thermal conductivity is obtained at near-critical density and temperature.

The theoretical treatment of the enhancement by means of the scaling theory shows the following.

- (a) The agreement between our experimental results and the theoretical approach by J. V. Sengers and co-workers is satisfactory.
- (b) There still remain some systematic deviations between measured and predicted data—a fact which has to be investigated. Measurements close to the critical density but far above the critical temperature may be useful to improve the correlation of background thermal conductivity at moderate densities.

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